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INTERNATIONAL CENTRE FOR
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LAMP
SERIES REPORT

(Laser, Atomic and Molecular Physics)

ULTRASONIC STUDY ON TERNARY LIQUID SYSTEMS BY LASER-SOUND INTERACTION

Mehdi Behboudnia

F. Necati Ecevit

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Ramazan Aydin

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ULTRASONIC STUDY ON TERNARY LIQUID SYSTEMS BY LASER–SOUND INTERACTION

Mehdi Behboudnia
Ostad Berzeger St., Kuhe 10, No.2, Urumiye, Iran,

F. Necati Ecevit
Department of Physics, Middle East Technical University,
06531 Ankara, Turkey

and

Ramazan Aydin [1]
International Centre for Theoretical Physics, Trieste, Italy.

ABSTRACT

To investigate the ultrasound velocity in liquid mixtures an interferometer based on Raman–Nath diffraction of laser light by sound waves is described. Ultrasonic velocity measurements in water in dependence of temperature and in carboxylic acids with triethylamine in benzene of different mole fractions are presented.

MIRAMARE – TRIESTE

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[1]Permanent address: Department of Physics, Middle East Technical University, 06531 Ankara, Turkey.

Preface

The ICTP-LAMP reports consist of manuscripts relevant to seminars and discussions held at ICTP in the field of Laser, Atomic and Molecular Physics (LAMP).

These reports aim at informing LAMP researchers on the activity carried out at ICTP in their field of interest, with the specific purpose of stimulating scientific contacts and collaboration of physicists from Third World Countries.

If you are interested in receiving additional information on the Laser and Optical Fibre activities at ICTP, kindly contact Professor Gallieno Denardo, ICTP.

ICTP LASER GROUP
P.O. BOX 586
34100 Trieste
Italy
Phone: +39+40+2240322
Fax: +39+40+224559
email: *passarel@ictp.trieste.it*

Introduction

The sound velocity in liquid mixtures is known to be a quantity which depends on concentration in a variety of manners according to the nature of each component liquid. Accurate measurements of ultrasonic velocity deliver information about physical and chemical behaviour of solutions and liquid mixtures such as molecular association and dissociation. Velocity values are also used to estimate several thermodynamical parameters such as adiabatic compressibility and molar volume that are, in turn, used to study intermolecular interactions in liquid mixtures.

Various theories and semiempirical models [1-5] are developed to compute ultrasonic velocity in liquid mixtures. The results are compared with experimental data and interpreted in terms of molecular interactions [6]. Flory's statistical theory [7], which utilises van der Waals potential energy model for obtaining the various configurational properties of liquids, has been modified by authors for predicting or directly calculating ultrasonic velocity in ternary liquid mixtures [8, 9].

The interaction of light with a transverse sound wave is described in terms of theories generally referred to as Raman-Nath diffraction or Bragg diffraction. If a beam of light is directed transversely through a sound wave, the angle of diffraction of the m^{th} order is given by the basic equation

$$\sin \theta_m = \pm m \lambda / \Lambda \qquad (1)$$

where λ is the wavelength of the incident light and Λ is the wavelength of sound in the medium. If the width of the sound beam, L, satisfies

$$2 \pi \lambda L / \Lambda^2 << 1 \qquad (2)$$

then "Raman-Nath" diffraction can be obtained under the right conditions and the incident light wave diffracts into multiple orders [10]. A detailed study of the Raman-Nath diffraction by ultrasonic waves is given in [11, 12].

This paper deals with an experimental study of ultrasonic velocity in liquids. A laser ultrasonic interferometer based on the appearance of Raman-Nath diffraction patterns under the laser illumination is described. The results of ultrasonic velocity measurements in water and in three different ternary liquid systems are presented.

Experimental

The interferometer employed in the experiments is illustrated in Fig. 1. The ultrasonic cell was a rectangular stainless steel vessel with transparent glass windows on either side. To produce the ultrasonic beam, a disk of piezoelectric ceramic 30 mm in diameter and 1.9 mm in thickness was used as a transducer, which was mounted on the cell very accurately. The movable metallic reflector (R) mounted on the opposite side serves to form standing sound waves in the cell. A

signal generator, LEADER Signal Generator Model 17A, was used as radio frequency (rf) source whose frequency change amounts to less than 3 kHz. It was adjusted to the second resonant frequency of 3.425 MHz of the transducer. The stability of the frequency was controlled by means of a B+k Precision Dynascan Co. 1805 frequency counter. The transmitting signal was amplified using a Model 420 LRF power amplifier of Electronic Navigation Industries. As light source we employed a He-Ne laser at the wavelength of 6328 A° and an Ar^+ laser at wavelengths of 4675 A° and 4880 A°. In order to produce the desired diffraction pattern, the width of the light beam must be larger than the wavelength of the sound. Therefore a collimator was used to widening the laser beam.

If D is the distance between the axis of the sound beam and the screen and d_m is the distance between m^{th} order and zeroth order diffraction pattern, since $D >> d_m$, it can be written that

$$\sin \Theta_m \cong d_m / D. \qquad (3)$$

Using the basic equation $\Lambda = V / \nu$, V and ν being the velocity and frequency of the sound wave respectively, we obtain from Eq.s (1) and (3)

$$V = (m \lambda \nu D) / d_m \qquad (4)$$

where d_m is measured by recording the diffraction pattern on a photograph.

The distance between the diffraction orders are only a few millimetres. For accurate measurement of these a camera with an exposure time of 1/1000 sec was used and the diffraction patterns were recorded on a film. The film was developed and all distances between the diffraction orders were measured with an accuracy of micrometers. A typical diffraction pattern photographed with the camera is shown in Fig. 2.

The performance of the arrangement was tested by measuring the ultrasound velocity in water at temperatures between 24 °C and 40 °C. The values obtained for various laser wavelengths are plotted versus temperature and presented in Fig.3.

We measured the ultrasonic velocity in carboxylic acids (acetic, propionic and butyric acids) with triethylamine (TEA) in benzene which form ternary liquid mixtures. Three systems, each containing five ternary samples, are examined. System I is composed of TEA (X1) + benzene (X2) + acetic acid (X3) while in systems II and III acetic acid is replaced by propionic acid and butyric acid, respectively. In each system the mole fraction of carboxylic acid is changed from 0.1 up to 0.5 at room temperature to form a total volume of 100 ml for each sample and the velocity of sound in each system is measured. All the measurements were made at constant temperature. Table 1 and Fig. 4 show the mole fractions of each system and the corresponding ultrasound velocities observed.

Table 1. The ultrasonic velocities measured for different mole fraction values of systems I, II and III at 22 ^{o}C.

(I): TEA (X1) + Benzene (X2) + Acetic acid (X3)		Velocity (m/s)
0.495	0.107	1233
0.395	0.197	1276
0.290	0.302	1325
0.198	0.404	1372
0.098	0.502	1338
(II): TEA (X1) + Benzene (X2) + Propionic acid (X3)		
0.507	0.099	1208
0.403	0.202	1252
0.301	0.304	1309
0.202	0.401	1347
0.102	0.500	1319
(III): TEA (X1) + Benzene (X2) +Butyric acid (X3)		
0.506	0.101	1235
0.402	0.202	1257
0.299	0.303	1314
0.200	0.403	1346
0.101	0.506	1318

Results and Discussion

As shown in Fig. 3, the measurements of sound velocity in water deliver, within the estimated experimental errors of 3 %, the same values for different wavelengths of laser light. This is a result of the relation of the wavelength of laser to the spacing of the diffraction orders as indicated in Eq. 4. The velocity of sound increases strongly as the temperature increases, which is a good agreement with the values given in literature, e.g., in [13]. It is well known that the speed of sound in pure water at room temperature increases with increasing temperature by about 3 m / s K, which is fairly satisfied in our results.

The results of sound velocity measurements in ternary liquids are presented in Table 1 and in Fig. 4. In all the systems of ternary mixtures, the ultrasonic velocity increases with the carboxylic acid content in the mixtures, reaches a maximum value at a critical concentration of 0.4 mole fraction of acid and then decreases with further increase of concentration. For comparison, calculated values [14] for the same liquids obtained at 30 ^{o}C are also shown in Fig. 4.

Using an inexpensive laser-sound interferometer we observed the Raman-Nath diffraction of laser light in various liquids and measured the speed of ultrasound waves. In conclusion we may say, that the results obtained in these experiments are in agreement with the available theoretical values given in [14] within the estimated systematic uncertainties. The results may be improved by carrying out the measurements at different temperatures, considering the temperature fluctuations and gradients, which were not taken into account in the current investigations.

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Figure Captions

Fig. 1. The experimental arrangement .

Fig. 2. A typical diffracted light beam pattern in water under Raman-Nath condition

Fig. 3. The change of ultrasonic velocity in water at temperatures between 24 °C and 40 °C for the wavelengths 476.5 nm and 488.0 nm of Ar ion laser and 632.8 nm of He-Ne laser.

Fig. 4. Plot of the sound velocity values versus the mole fraction of carboxylic acids: (a) experimental at 22 °C (this work) and (b) calculated literature values at 30 °C [14].

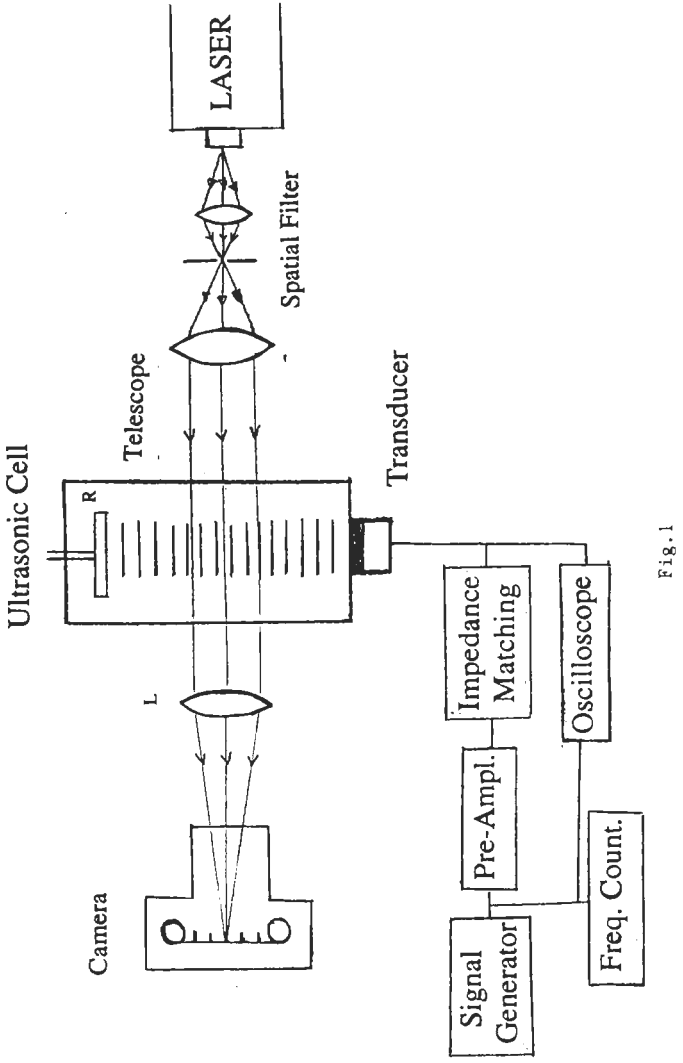

Fig.1

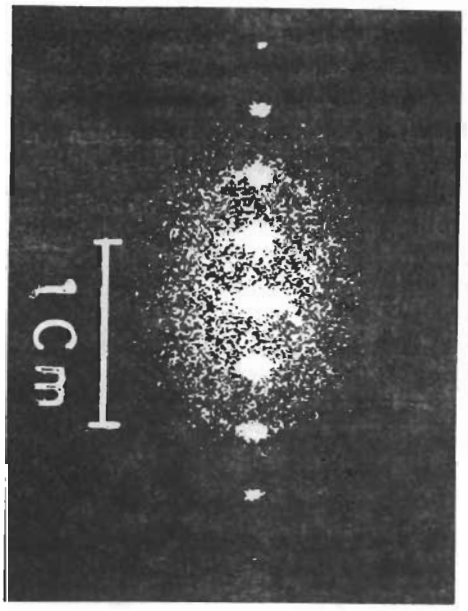
1 Cm

Fig.2

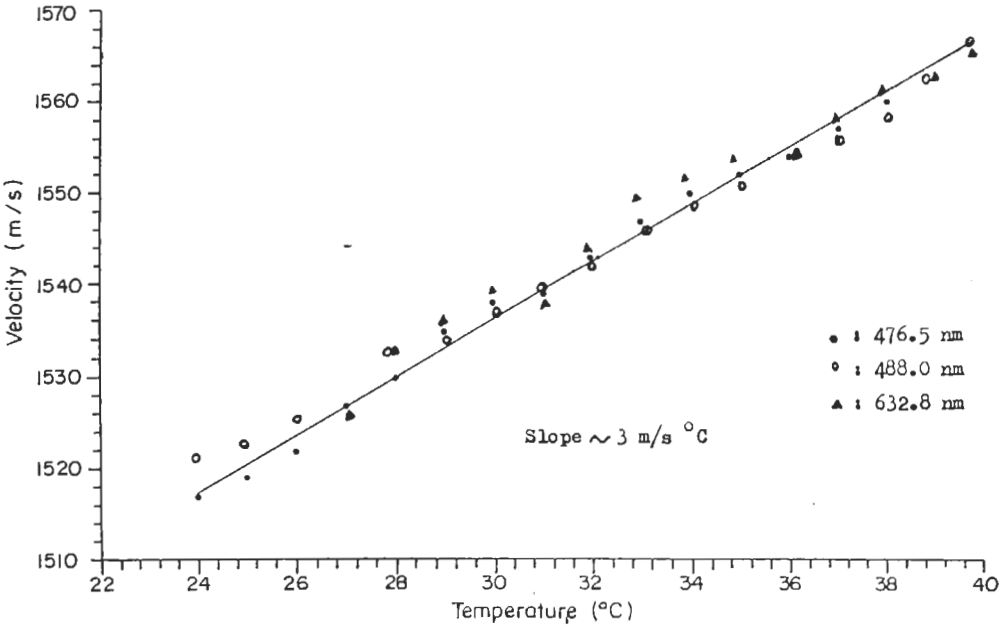
1570
1560
1550
1540
1530
1520
1510
Velocity (m/s)
22
24
26
28
30
32
34
36
38
40
Temperature (°C)
• : 476.5 nm
° : 488.0 nm
▲ : 632.8 nm
Slope ~ 3 m/s °C

Fig.3

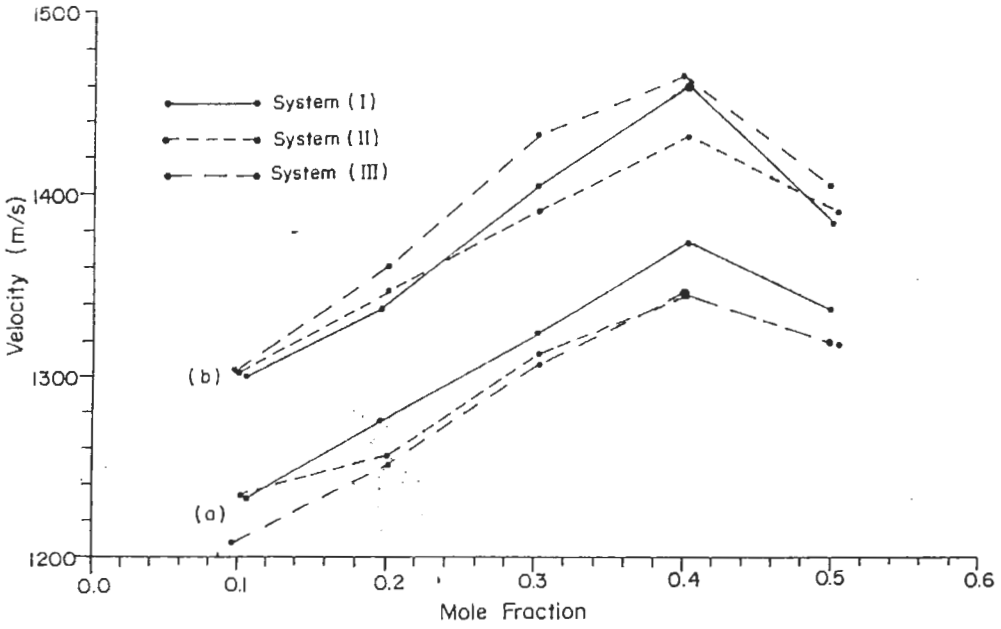
1500
1400
1300
1200
Velocity (m/s)
System (I)
System (II)
System (III)
(b)
(a)
0.0
0.1
0.2
0.3
0.4
0.5
0.6
Mole Fraction

Fig.4